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Re-Measured Uranium Branching Ratios and their Impact on Removing Biases from MGAU Analyses

T-F. Wang, W.D. Ruhter, R.G. Lanier

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Re-Measured Uranium Branching Ratios and their Impact on Removing Biases from MGAU Analyses

Tzu-Fang Wang, Wayne D. Ruhter, and Robert G. Lanier
Lawrence Livermore National Laboratory
Livermore, CA 94550

Abstract

Biases in MGAU analyses first observed in FSU Gosatomnadzor inspection and subsequently identified in more detail by measurements at the Moscow Kurchatov Institute have forced a new look at the code's analysis assumptions.

We have used uranium gamma-ray calibration standards from the National Bureau of Standards and standards from the New Brunswick Laboratory to investigate MGAU analysis biases. The 200g uranium standards which cover the uranium enrichments ranging from 0.3% to 93% were used to collect more than 500 gamma-ray spectra for this study. The experimental arrangement used a LEPS Ge detector with various source-detector configurations and absorbers. Two independent versions of the MGAU code, which we currently employ in our laboratory, confirm the biases noted in a developing variety of FSU inspection results and in the careful Kurchatov study.

In this paper, we will discuss the MGAU methodology and use 250 spectra at a fixed geometry without absorbers to obtain new branching ratios for the critical 100-keV region gamma rays. We show that modifying the branching ratios removes a significant component of the observed biases.

Introduction

Gamma ray spectrometry using high-purity Ge detectors has been a powerful tool for non-destructive evaluation of plutonium assays. Isotopic ratios can be obtained by comparing the strength of the gamma rays from the decay of each nucleus. There are several widely used isotopic analysis codes such as MGA[1] from Lawrence Livermore National Laboratory (LLNL) and FRAM[2] from the Los Alamos National Laboratory (LANL) which successfully employ such analysis algorithms.

In the past few years, new isotopic analysis codes such as CZTU [3] and MGAU [4] have been developed at LLNL for dealing with uranium accountability issues. MGAU is based on the MGA methodology and performs spectral de-convolution of the complicated 100-keV gamma- and x-ray region of uranium spectra. Unlike MGA, which de-convolutes plutonium 100-keV spectra based on the Pu-Am gamma rays, MGAU relies on the ^{235}U and ^{238}U daughter gamma rays of, Th and Pa. For the analysis to be accurate – i.e. for the code to calculate an accurate $^{235}U/^{238}U$ isotopic ratio for the sample - ^{235}U and ^{238}U must be in equilibrium with their short-lived (less than a month half-life) daughters.

Recent information from FSU field inspections and from a more detailed study[5] of MGAU performance

at the Moscow Kurchatov Institute, it has become apparent that biases very likely exist in such measurements. In order to investigate this problem carefully, we undertook our own measurements under strictly controlled conditions. We used certified 200g uranium gamma-ray standards provided by the National Bureau of Standards and by the New Brunswick Laboratory and confirmed that our versions of the MGAU codes show similar biases when de-convoluting the spectra from these samples.

MGAU Methodology and Problems

MGAU de-convolutes the complicated 100 keV gamma- and x-ray region of a "Uranium" spectrum. Figure 1 shows an overlay of three spectra in this energy region and representing various levels of ^{235}U enrichment: (3%, 52% and 0.7%. The 92.798 keV and 92.385 keV gamma rays are a close lying doublet and contain the information about ^{238}U enrichment in the sample. When the ^{235}U enrichment is low, this doublet, as well as the $K_{\alpha 1}$ and $K_{\alpha 2}$ uranium x-rays, will dominate this region. For highly enriched uranium, the doublet is hidden under the "^{235}U" gamma rays and under the tail of uranium x rays. In the range of 10% ^{235}U enrichment, the doublet height will be roughly as high as the nearby "^{235}U" gamma rays.

The algorithm for determining background in MGAU in the 100-keV region is identical to the background determination algorithm in MGA and is described in detail in ref. 1 and in ref. 6. Basically speaking, the background subtraction algorithm relies on the height of the spectra. This integrative step-function-like background-determination algorithm, although it can not represent the real background, is probably the closest to reality among various background subtraction algorithms [6]. The gamma rays are de-convoluted using Gaussian plus tails functions, and the x-rays are unfolded using Voigt functions. Multi-group fittings were use.

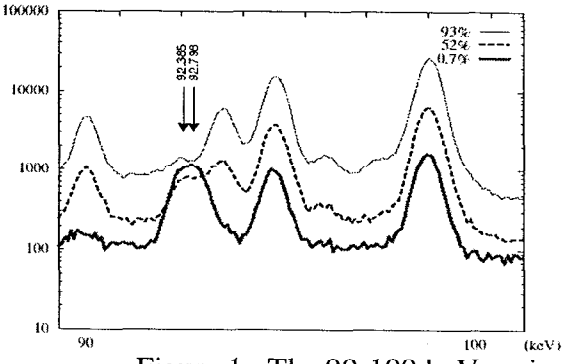


Figure 1. The 90-100 keV region of uranium gamma-ray spectrum. The ^{238}U doublet is shown. The two large peaks in the middle and to the right in the 0.7% spectrum are the uranium $K_{\alpha 2}$ and $K_{\alpha 1}$ x rays, respectively.

Experimental Setup

A low-energy high-purity germanium detector (LEPS) was used to detect 100-keV gamma rays from the daughters of ^{235}U and ^{238}U. A spectroscopic amplifier and a 4K-channel ADC were used to obtain the gamma-ray data to 300 keV. The system gain was operated at the "MGAU-specified" value of .075keV/channel. Five National Bureau of Standards samples (0.31%, 0.71%, 1.94%, 2.95%, and 4.46%) were used to cover the low ^{235}U enriched regime while three standards from the New Brunswick Laboratory were used to study the medium-to-high enriched regime (20.06%, 52.56%, and 93.18%). Approximately 30 spectra were collected for each standard. The sources were

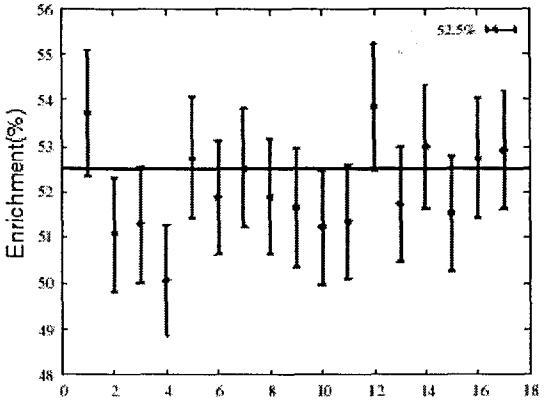
place 15 cm away from the detector and a one-inch diameter collimator was placed in front of the source. Different collecting times were used for each source and ranged from thirty minutes to two hours. The detector resolution was approximately 540eV for 120keV gamma rays. A 2μS shaping time was used and the dead time was below 3% for all the measurements.

The branching ratios

The widely varying spectral features, which are encountered over the range of enrichments that one hopes to apply MGAU, presents a challenging spectroscopy problem. Moreover, the nature of the algorithms that perform, the de-convolutions are sensitive to instrument parameters. For these reasons, it is not easy to uniquely identify general biases in the uranium assay analysis under ordinary usage. In the present measurements we worked to eliminate these obvious sources of bias and determined that a residual bias still existed. For this reason we undertook a re-evaluation of the important uranium branching ratios.

It is hard to determine the branching ratios for gamma rays in the 100keV energy region because of x-ray tails and uncertainties in applying the background subtraction algorithm to remove their affect from the measurement. However, by using thin uranium targets the x-ray fluorescence can be reduced. As a result, the desired data is less obscured by x-ray contamination and therefore the analyses are less dependent on background compensation. The measurements are difficult; they require a long counting time and data must be accumulated in an extremely low background environment. This is likely why there are large discrepancies among the compiled branching ratios [7,8] for this energy region. In most of the cases, these branching ratios are even not within the quoted errors.

To obtain better branching ratio values, we used low enriched spectra to determine the energy and the branching ratios of the 92.798, 92.385 keV doublet. By contrast and to take advantage of better counting statistics, we used the high enriched spectra to determine the energy and branching ratios of the "^{235}U" gamma rays. By applying this new set of energy and branching ratio data, the average enrichment values obtained from MGAU are within 1% of their standard values throughout the entire collected uranium enrichment spectra. Figure 2 shows MGAU results of two sets of high-enrichment data compared to their standard values. It is worth noting that the ^{234}U value derived from the 120.9 keV gamma ray is also in good agreements with the standard values for the three high-enriched standards.



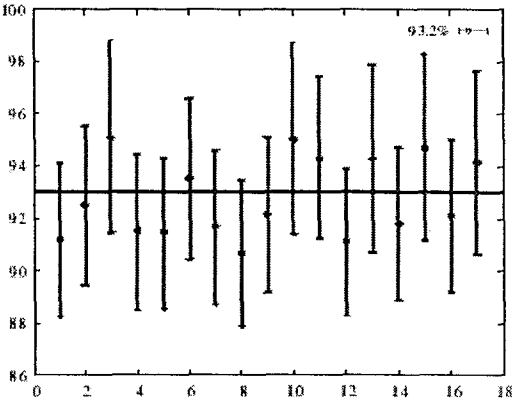


Figure 2. The results from MGAU with the new set of energies and branching ratios, the black line in each graph represent the standard enrichment value of the source. The averaged bias was – 0.98% for 52.5% enrichment data and – 0.21% for 93.2% enrichment data. The old MGAU results show +3.1% for 52.5% data and +2.3% for 93.2% data. All 18 spectra for each standard are collected under the same geometry with the same live time.

Conclusion

Using data from the uranium gamma-ray standards, we have derived a set of self-contained energy-and-branching ratio of the 100-keV region gamma rays for MGAU. There are other problems in MGAU such as corrections due to absorbers. However these problems will be addressed at a later date.

Acknowledgement

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